Supporting Information

All reactions were carried out under an argon atmosphere in oven dried glassware unless otherwise specified. Ethyl acetate was purchased anhydrous from Aldrich Chemical Company and was used without further purification. Methanol was distilled under a nitrogen atmosphere over calcium hydride. Triethylamine was distilled over calcium hydride and stored over potassium hydroxide. Formic acid was purchased from Breckland Scientific Supplies. Pd-EnCatTM was obtained from Avecia. All other reagents were used as supplied by Aldrich, Acros or Lancaster. NMR spectra were recorded in CDCl₃ unless otherwise stated. NMR spectra were recorded at 25 °C in 5mm tubes, at 400.1 MHz (1 H) or 100.6 MHz (13 C) on a Bruker DPX400 instrument. Chemical shifts are quoted relative to the residual solvent peak (7.24 δ for 1 H and 77.0 δ for 13 C). IR spectra were recorded using Perkin-Elmer Spectrum One FT-IR spectrometer as neat solids. Mass spectra were recorded on a Kratos 890 spectrometer. Optical rotations were recorded on a Perkin-Elmer Model 343 Polarimeter.

Cis-2-methyl-3-phenyl-oxirane¹

Cis-β-methyl styrene (0.23 g, 1.95 mmol) was added portionwise to a stirred solution of *m*-chloroperbenzoic acid (77 %, 0.56 g, 2.5 mmol) in dichloromethane (30 mL). The reaction mixture was stirred at room temperature for 14 hours. Calcium hydroxide (0.25g) was added, and the reaction mixture filtered and concentrated under reduced pressure to afford 0.22g (84 %) of the epoxide as a clear, colourless oil: ¹H NMR (400 MHz, CDCl₃) δ 7.38-7.30 (m, 5H), 4.07 (d, J 4.3 Hz, 1H), 3.35 (dq, J 4.3, 5.5 Hz, 1H), 1.10 (d, J 5.5 Hz, 3H); ¹³C NMR (100 MHz, CDCl₃) δ 135.5, 128.0, 127.5, 126.6, 57.5, 55.1, 12.5; spectra consistent with known data. ¹

1,2-epoxy-2-phenylcyclohexene²

1-phenyl-1-cyclohexene (0.53 mL, 0.53 g, 3.16 mmol) was added portionwise to a stirred solution of m-chloroperbenzoic acid (77 %, 0.84 g, 3.75 mmol) in dichloromethane (15 mL) at 0 °C. The reaction mixture was stirred at room temperature for 18 hours. The reaction mixture was diluted with 10 % NaOH (50 mL), extracted with dichloromethane (3 x 30 mL), dried with MgSO₄, filtered and concentrated under reduced pressure. Purification by column chromatography (2 % Et₂O in pet. ether) afforded 0.36g (66 %) of 1,2-epoxy-2-phenylcyclohexene as a clear, colourless oil: 1 H NMR (400 MHz, CDCl₃) δ 7.39-7.27 (5H, m), 3.07 (m, 1H), 2.35-1.95 (m, 4H), 1.68-1.18 (m, 4H); 13 C NMR (100 MHz, CDCl₃) δ 142.5, 128.2, 127.2, 125.3, 61.9, 60.2, 28.9, 24.7, 20.1, 19.8; IR (neat) v 3028, 2936, 2860, 1494, 1446; m/z (EI) 174.1036 (M $^{+}$ - C₁₂H₁₄O requires 174.1045).

1,2-epoxy-2-phenyl-cyclopentene³

1-phenyl-1-cyclopentene (0.47 g, 3.18 mmol) was added portionwise to a stirred solution of *m*-chloroperbenzoic acid (77 %, 0.89 g, 3.97 mmol) in dichloromethane (15 mL) at 0 $^{\circ}$ C. The reaction mixture was stirred at room temperature for 14 hours. The reaction mixture was diluted with 10 % NaOH (50 mL), extracted with dichloromethane (3 x 30 mL), dried with MgSO₄, filtered and concentrated under reduced pressure. Purification by column chromatography (5 % Et₂O in pet. ether) afforded 0.27g (55 %) of 1,2-epoxy-2-phenyl-cyclopentene as a clear, colourless oil: 1 H NMR (400 MHz, CDCl₃) δ 7.42-7.28 (m, 5H), 3.58 (m, 1H), 2.23-1.58 (m, 6H, m); 13 C NMR (100 MHz, CDCl₃) δ 138.0, 128.3, 127.6, 125.9, 66.9, 66.4, 28.9, 28.1, 19.3; IR (neat) v 3029, 2954, 2854, 1604, 1498, 1403; m/z (EI) 160.0882 (M $^{+}$ - C₁₁H₁₂O requires 160.0889).

1-(3-phenyl-oxiranyl)-ethanone ⁴

Hydrogen peroxide (35 %, 1 mL, 11.7 mmol) was added slowly over 10 minutes to a stirred solution of *trans*-4-phenyl-buten-3-one (0.52g, 3.6 mmol) in methanol (5 mL) at 5

°C. NaOH (2M, 1 mL) was added over 20 minutes, and the mixture stirred at room temperature for 3 hours. Saturated Na₂S₂O₅ solution was used to destroy any remaining peroxide whilst keeping the temperature below 40 °C. The mixture was diluted with water (40 mL), extracted with ether (3 x 40 mL), dried with MgSO₄, filtered and concentrated under reduced pressure. Purification by column chromatography (9:1 pet. ether: Et₂O) yielded 0.31g (54%) of 1-(3-phenyl-oxiranyl)-ethanone as a white solid: mp 41-42°C (lit.⁵ mp 40-41.5 °C (MeOH)); ¹H NMR (400 MHz, CDCl₃) δ 7.42-7.21 (m, 5H), 4.00 (d, *J* 1.8 Hz, 1H), 3.48 (d, *J* 1.8 Hz, 1H), 2.18 (s, 3H); ¹³C NMR (100 MHz, CDCl₃) δ 204.0, 135.0, 128.9, 128.6, 125.6, 63.4, 57.6, 24.7; IR (neat) v 3064, 3013, 1708, 1460, 1355; m/z (ESI) 185.0577 (M⁺ + Na - C₁₀H₁₀O₂ Na requires 185.0579).

1-(3-pentyl-oxiranyl)-ethanone

Hydrogen peroxide (35 %, 1.2 mL, 14.1 mmol) was added slowly over 10 minutes to a stirred solution of *trans*-3-nonen-2-one (0.43 mL, 0.35g, 2.47 mmol) in methanol (5 mL) at 5 °C. NaOH (2M, 1 mL) was added over 30 minutes, and the mixture stirred at room temperature for 7 hours. Saturated Na₂S₂O₅ solution was used to destroy any remaining peroxide whilst maintaining the temperature below 40 °C. The mixture was diluted with water (40 mL), extracted with ether (3 x 40 mL), dried with MgSO₄, filtered and concentrated under reduced pressure. Purification by column chromatography (9:1 pet. ether: Et₂O) yielded 0.24g (61 %) of 1-(3-pentyl-oxiranyl)-ethanone as a clear, colourless oil: ¹H NMR (400 MHz, CDCl₃) δ 3.16 (d, *J* 2.0 Hz, 1H), 3.06 (td, *J* 5.5, 2.0 Hz,1H), 2.04 (s, 3H), 1.61 (m, 2H), 1.45 (m, 2H), 1.32 (m, 4H), 0.88 (t, *J* 7.0 Hz, 3H); ¹³C NMR (100 MHz, CDCl₃) δ 206.0, 59.9, 58.0, 31.7, 31.6, 25.4, 24.3, 22.4, 13.9; IR (neat) v 2930, 2860, 1709; m/z (ESI) 179.1048 (M⁺ + Na – C₉H₁₆O₂ Na requires 179.1049).

Preparation of EnPd(OAc)₂ (Pd-EnCatTM)

EnPd(OAc)₂ was prepared by Avecia using a procedure based on the following protocol: polymethylene polyphenylene di-isocyanate (SUPRASEC 5025, average functionality of

2.7) and the palladium acetate was dissolved in chloroform. This solution was then dispersed at 800rpm for one minute into an aqueous solution of sodium lignosulfonate (Reax 100 M), polyvinyl alcohol (Gohsenol GL 03) and the polyoxyethylene ether of butyl alcohol (Tergitol XD) using a laboratory overhead stirrer fitted with a 50mm rotary blade. The resulting oil in water emulsion had a particle size range of 20-250 microns and was gently shaken for 16h. The polyurea microcapsules obtained were filtered through a polyethylene frit (20-micron porosity), and were washed with de-ionized water (5 x 50mL), aceteone (5 x 50mL) and ether (3 x 50mL) and dried at room temperature.

Reduction of EnPd(OAc)₂ to Pd⁰-EnCatTM

EnPd(OAc)₂ (0.4 mmol/g, 0.4 g) in ether (8 mL) and formic acid (8 mL) at 45 °C for 2 hours. The mixture was allowed to cool, and the microcapsules filtered through a polyethylene frit (20 micron porosity), washed with distilled water (3 x 30mL), acetone (3 x 30mL) and ether (3 x 30mL). The microcapsules were dried under oil pump vacuum (approximately 0.5 mm Hg or better) at 45 °C for 5 hours to yield black Pd⁰-EnCat microcapsules (0.35g).

General procedure for reductive cleavage of epoxides by transfer hydrogenation

The epoxide substrate (0.5 mmol, 0.4 M) and Pd⁰-EnCatTM (0.4 mmol/g, 63 mg, 0.025 mmol) were dissolved in anhydrous ethyl acetate (0.75 mL). Distilled triethylamine (0.30 mL, 0.22 g, 2.15 mmol) and then formic acid (90 %, 0.09 mL, 100mg, 2.15 mmol) were added, and the reaction stirred under argon at 23 °C until TLC analysis indicated that the reaction was complete. The reaction mixture was decanted off, and the microcapsules washed with ethyl acetate. The mixture was concentrated under reduced pressure and the residue purified by flash column chromatography (eluting with ether in pet. ether) to yield the alcohol product, details of any other products are given if sufficient purified material was obtained for analysis.

2-phenyl ethanol ⁶

Purification by column chromatography (2:1 pet. ether: Et₂O) yielded 400mg (84 %) of the alcohol as colourless oil: 1 H NMR (400 MHz, CDCl₃) δ 7.22-7.34 (m, 5H), 3.87 (t, *J* 6.6 Hz, 2H), 2.88 (t, *J* 6.6 Hz, 2H), 1.43 (s, 1H); 1 C NMR (100 MHz, CDCl₃) δ 138.5, 129.0, 128.5, 126.4, 63.6, 39.1; IR (neat) v 3300), 3063, 3028, 2944, 2874, 1603, 1496, 1453; m/z (EI) 122.0733 (M⁺– C₈H₁₀O₁ requires 122.0732).

2-Phenylethyl formate

¹H NMR (400 MHz, CDCl₃) δ 8.04 (s, 1H), 7.33-7.23 (m, 5H), 4.41 (t, *J* 7.0 Hz, 2H), 2.99 (t, *J* 7.0 Hz, 2H); ¹³C NMR (100 MHz, CDCl₃) δ 160.1, 137.8, 128.5, 128.4, 126.7, 64.3, 34.9; spectra consistent with known data.⁷

(R)-(-)-1-phenyl-propan-2-ol

Purification by column chromatography (2:1 pet. ether: Et₂O) yielded 45.6 mg (90 %) of the alcohol as colourless oil: $[\alpha]_D^{25}$ -32.8° (c=1.025, CHCl₃)⁸; ¹H NMR (400 MHz, CDCl₃) δ 7.35-7.21 (m, 5H), 4.01 (m, 1H), 2.78 (dd, *J* 13.4, 5.0 Hz, 1H), 2.70 (dd, *J* 7.8, 13.4 Hz, 1H), 2.01 (br s,1H), 1.25 (d, *J* 6.2 Hz, 3H); ¹³C NMR (100 MHz, CDCl₃) δ 138.5, 129.4, 128.5, 126.5, 68.8, 45.8, 22.8; IR (neat) v 3350, 3062, 3027, 2970, 2029, 1599, 1496, 1453; m/z (EI) 136.0895 (M⁺– C₉H₁₂O₁ requires 136.0889).

1-phenylpropan-2-one

 1 H NMR (400 MHz, CDCl₃) δ 7.34-7.20 (m, 5H), 3.70 (s, 2H), 2.16 (s, 3H); spectra consistent with known data. 9

1,2-diphenyl-ethanol 10

Purification by column chromatography (2:1 pet. ether: Et₂O) yielded 95.3 mg (99%) of the alcohol as yellow crystalline solid: mp 65-66 °C (lit.¹⁰ mp. 66 °C); ¹H NMR (400 MHz, CDCl₃) δ 7.38-7.20 (m, 10H), 4.92-4.89 (m, 1H), 3.06-3.02 (m, 2H), 2.00 (s, 1H); ¹³C NMR (100 MHz, CDCl₃) δ 143.8, 138.0, 129.5, 128.5, 128.4, 127.6, 126.6, 125.9, 75.3, 46.1; IR (neat) v 3380, 3062, 3028, 2918, 1602, 1494, 1453; m/z (EI) 198.1036 (M⁺– C₁₄H₁₄O₁ requires 198.1045).

3-phenyl-propan-1,2-diol 11

Purification by column chromatography (2:1 pet. ether: Et₂O) yielded 72.5 mg (95 %) of the alcohol as white solid; mp 64-65 °C; ¹H NMR (400 MHz, CDCl₃) δ 7.20-7.33 (m, 5H), 3.91 (d, *J* 7.2 Hz, 1H), 3.65 (d, *J* 11.1 Hz, 1H), 3.65 (d, *J* 11.1 Hz, 1H), 3.45-3.50 (s, 1H), 2.76 (d, *J* 7.4 Hz, 2H), 2.56 (br s, 2H); ¹³C NMR (100 MHz, CDCl₃) δ 137.8, 129.3, 128.6, 126.5, 73.0, 66.0, 39.8; IR (neat) v 3230, 3029, 2923, 2862, 1495, 1456; m/z (EI) 152.0842 (M⁺– C₉H₁₂O₂ requires 152.0838).

Methyl-3-phenyl-propan-1,2-diol 12

Purification by column chromatography (Et₂O) yielded 63.6mg (93 %) of the alcohol as white crystals: mp 65-67 °C (lit.¹³ mp.66.5-67.5 °C ¹H NMR (400 MHz, CDCl₃) δ 7.44-7.34 (m, 5H), 3.60 (d, *J* 10.9 Hz, 1H), 3.54 (d, *J* 10.9 Hz, 1H), 2.96 (d, *J* 13.3 Hz 1H), 2.90 (d, *J* 13.3 Hz, 1H), 2.07 (br s, 2H), 1.25 (s, 3H); ¹³C NMR (100 MHz, CDCl₃) δ

137.0, 130.4, 128.3, 126.5, 73.3, 69.2, 44.6, 23.5;); IR (neat) v 3359, 3028, 2971, 2926, 1602, 1494, 1453; m/z (ESI) 189.0890 (M^+ + Na– $C_{10}H_{14}O_2Na$ requires 189.0892).

2-Hydroxy-3-phenyl-propionic acid ethyl ester 14

Purification by column chromatography (1:2 pet ether : Et₂O) yielded 89.5 mg (92 %) of the alcohol as a clear liquid: 1 H NMR (400 MHz, CDCl₃) δ 7.33-7.22 (m, 5H), 4.44 (m, 1H) 4.22 (q, J 7.1 Hz, 2H), 3.13 (dd, J 4.5, 13.9 Hz, 1H), 2.97 (dd, J 6.8, 13.9 Hz, 1H), 2.84 (d, J 6.2 Hz, 1H), 1.28 (t, J 7.1 Hz, 3H); 13 C NMR (100 MHz, CDCl₃) δ 174.6, 136.3, 129.5, 128.3, 126.8, 71.2, 61.6, 40.5, 14.1; IR (neat) v 3480, 3030, 2980, 1730 (s), 1497, 1456; m/z (ESI) 217.0842 (M⁺ + Na – C₁₁H₁₄O₃Na requires 217.0841).

2-Hydroxy-3-phenyl-butyric acid ethyl ester

The starting material used was a mixture of cis and trans ethyl 3-methyl-3-phenylglycidate. Purification by column chromatography (2:1 pet ether : Et₂O) yielded 99.4 mg (94 %) of the alcohol as a clear liquid, as a one to one mixture of diastereomers: 1 H NMR (400 MHz, CDCl₃) δ 7.21-7.32 (m, 10H), 4.31 (br s, 2H), 4.20 (q, J 7.1 Hz, 2H), 4.14 (q, J 7.2 Hz, 2H), 3.28-3.25 (m, 1H), 3.24-3.20 (m, 1H), 2.85 (d, J 6 Hz, 1H), 2.68 (d, J 6 Hz, 1H), 1.45 (d, J 7.2 Hz, 3H), 1.37 (d, J 7.1 Hz, 3H), 1.24 (t, J 7.2 Hz, 3H), 1.23 (t, J 7.1 Hz, 3H); 13 C NMR (100 MHz, CDCl₃) δ 174.1, 173.7, 142.6, 140.7, 128.3, 128.2, 127.8, 126.7, 75.0, 74.9, 61.6, 61.5, 43.4, 17.6, 14.6, 14.1, 14.0; IR (neat) v 3490, 3028, 2980, 2936, 1726, 1453; m/z (ESI) 231.0993 (M⁺ + Na - C₁₂H₁₆O₃Na requires 231.0997).

2-(4-Fluoro-phenyl)-ethanol 15

Purification by column chromatography (1:1 pet ether : Et₂O) yielded 58.0mg (82 %) of the alcohol as a yellow oil: 1 H NMR (400 MHz, CDCl₃) δ 7.10-6.88 (m, 4H), 3.72 (d, J 6.6 Hz, 2H), 2.73 (d, J 6.6 Hz, 2H), 2.31 (br s, 1H); 13 C NMR (100 MHz, CDCl₃) δ 161.6 (d, 1 J_{CF} 242.8 Hz), 134.1 (d, 4 J_{CF} 3.3 Hz), 130.3 (d, 3 J_{CF} 7.9 Hz), 115.2 (d, 1 J_{CF} 21.0 Hz), 63.5, 38.2; IR (neat) v 3340, 2937, 2875, 1721, 1601, 1508, 1415; m/z (EI) 140.06375 (M $^{+}$ – C₈FH₉O requires 140.06377).

2-Hydroxy-3-(4-methoxy-phenyl)-propionic acid methyl ester 16

Purification by column chromatography (3:1 pet ether : Et₂O) yielded 58.0 mg (82 %) the alcohol as a yellow oil: 1 H NMR (400 MHz, CDCl₃) δ 7.13-6.82 (m, 4H), 4.41 (m, 1H), 3.78 (s, 3H), 3.76 (s, 3H), 3.06 (dd, *J* 4.4 Hz, 14.0 Hz, 1H), 2.91 (dd, *J* 6.6 Hz, 14.0 Hz, 1H), 2.74 (br s, 1H); 13 C NMR (100 MHz, CDCl₃) δ 174.5, 158.6, 130.5, 128.2, 113.9, 71.4, 55.2, 52.4, 39.6; IR (neat) v 3475, 2955, 2837, 1733, 1611, 1511; m/z (ESI) 233.0792 (M⁺ + Na– C₁₁H₁₄O₄Na requires 233.0790).

3-Hydroxy-4-phenyl-butan-2-one (1) and 3-hydroxy-1-phenyl-butan-2-one (2) 17

Purification by column chromatography (1:1 pet ether : Et₂O) yielded 71.0 mg (87 %) of the product as a clear oil an inseparable mixture of products **1** and **2** in a 6:1 ratio: 1 H NMR (400 MHz, CDCl₃) δ 7.36-7.20 (m, 5H), 4.41 (**1**, dd, *J* 4.7, 7.3 Hz, 1H), 4.34 (**2**, q, *J* 7.0 Hz, 1H), 3.82 (**2**, d, *J* 15.8 Hz, 1H), 3.76 (**2**, d, *J* 15.8 Hz, 1H), 3.5 (br, OH), 3.13 (**1**, dd, *J* 5.3, 14.2 Hz, 1H), 2.88 (**1**, dd, *J* 5.3 Hz, 14.2 Hz, 1H), 2.19 (**1**, s, 3H), 1.41 (**2**, d, *J* 7.0 Hz, 3H); 13 C NMR (100 MHz, CDCl₃) δ 209.9 (**2**), 209.2 (**1**), 136.4 (**1**), 133.0 (**2**), 129.3 (**2**), 129.2 (**1**), 128.7 (**2**), 128.5 (**1**), 127.2 (**2**), 126.8 (**1**), 77.6 (**1**), 72.2 (**2**), 44.5 (**2**), 39.8 (**1**), 25.8 (**1**), 19.7 (**2**); IR (neat) v 3415, 3030, 2870, 2251, 1711, 1603, 1454; m/z (ESI) 187.0740 (M⁺ + Na- C₁₀H₁₂O₂ requires 187.0735).

cis-2-Phenyl-cyclohexanol

Purification by column chromatography (9:1 pet ether : Et₂O) yielded 72.2 mg (84 %) of the alcohol as a yellow solid: mp 41-43 °C; ¹H NMR (400 MHz, CDCl₃) δ 7.36-7.22 (m, 5H), 4.02 (s, 1H), 2.76 (d, *J* 12.8 Hz), 2.08 (dq, *J* 3.6, 12.8 Hz, 1H), 2.01-1.97 (m, 1H), 1.94-1.33 (m, 6H); ¹³C NMR (100 MHz, CDCl₃) δ 144.0, 128.9, 127.7, 126.4, 70.5, 48.0, 32.9, 26.2, 24.3, 19.5; IR (neat) v 3440, 3026, 2929, 2860, 1601, 1496, 1446; m/z (EI) 176.1195 (M⁺– C₁₂H₁₆O requires 176.1202); product identified by comparison to spectra for *trans*-2-phenyl-cyclohexanol. ¹⁸

3-hydroxy-1-cyclohexanone 19

Purification by column chromatography (Et₂O) yielded 45.4 mg (81 %) of the alcohol as a colourless oil: 1 H NMR (400 MHz, CDCl₃) δ 4.18 (br s, 1H), 2.64 (dd, J 4.1 Hz, 14.0 Hz, 1H), 2.39 (dd, J 7.6, 14.0 Hz, 1H), 2.30 (t, J 6.5 Hz, 2H), 2.24 (s, 1H), 2.11-1.96 (m, 2H), 1.80-1.64 (m, 2H); 13 C NMR (100 MHz, CDCl₃) δ 210.0, 69.7, 50.4, 40.9, 32.8, 20.6; IR (neat) ν 3540, 2940, 2300, 2250, 1712; m/z (EI) 114.0679 (M $^{+}$ – C₆H₁₀O₂ requires 114.0681).

Reduction of 2-benzyl oxirane

2-benzyl oxirane (0.20g, 1.52 mmol) and Pd⁰-EnCatTM (0.4 mmol/g, 191 mg, 0.076 mmol) were dissolved in distilled methanol (3 mL). The reaction was stirred under hydrogen (40 bar) for 19 hours. The reaction mixture was decanted off, and the microcapsules washed with ethyl acetate. The mixture was concentrated under reduced

pressure and the residue purified by flash column chromatography (2:1 Et₂O : pet ether), yielding 144.2 mg (70 %) of 1-phenyl-propan-2-ol (**3**) as a colourless oil: 1 H NMR (400 MHz, CDCl₃) δ 7.35-7.21 (m, 5H), 4.01 (m, 1H), 2.78 (dd, J 13.4, 5.2 Hz, 1H), 2.70 (dd, J 7.8, 13.4 Hz, 1H), 2.01 (br s,1H), 1.25 (d, J 6.2 Hz, 3H); 13 C NMR (100 MHz, CDCl₃) δ 138.5, 129.4, 128.5, 126.5, 68.8, 45.8, 22.8; IR (neat) v 3350, 3062, 3027, 2970, 2029, 1599, 1496, 1453; m/z (EI) 136.0895 (M⁺– C₉H₁₂O₁ requires 136.0889); and 29.7 mg (15%) of 3-phenyl-propan-1-ol (**4**) as a clear colourless oil: 1 H NMR (400 MHz, CDCl₃) δ 7.35-7.21 (m, 5H), 3.64 (t, J 5.7 Hz, 2H), 2.72 (t, J 5.7 Hz, 2H), 1.99 (s, br, 1H), 1.89 (m, 2H); 13 C NMR (100 MHz, CDCl₃) δ 141.8, 128.4, 125.9, 62.3, 34.2, 32.1; spectra consistent with known data. 20

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